Integration of Multi-Stage Membrane Carbon Capture Processes to Coal-Fired Power Plants using highly permeable polymers

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Research paper

Integration of multi-stage membrane carbon capture processes to coal-fired power plants using highly permeable polymers

Maria-Chiara Ferrari*, Davide Bocciardo, Stefano Brandani

School of Engineering, University of Edinburgh, The King's Buildings, Robert Stevenson Road, EH9 3FB, Edinburgh, UK

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Abstract

Membrane separation systems could be a feasible option as post combustion carbon capture technologies in coal-fired power plants. Recent advancement on membrane materials based on microporous super glassy polymers could improve significantly the capture process but the properties of the materials have to guide the design of the separation stage. In this study an advanced hybrid two-stage membrane process employing one of the most permeable polymer known (PIM-1) is retrofitted to a coal fired power plant and the process is analysed in terms of energy requirement and cost performance. The results are based on the use of an in-house detailed membrane module model implemented in UniSim Design®, the Honeywell process flowsheet simulator. The study indicates the need for advanced configuration in order for highly permeable membranes to be competitive with more mature technologies in terms of capture cost. The effect of ageing and impurities on the material is also investigated in order to predict the decline in process performance over time and suggest a timeproof design.

Keywords: Membranes; PIMs; Post-combustion; LCOE

1. Introduction and background

Carbon dioxide emissions from power plants are one of the main contributors to global climate change [1] and significant efforts in their reduction are needed if the increase in global average temperature is to be limited well below 2 °C as pledged at COP21 in December 2015 in Paris [2]. Combustion of fossil fuels will still play a key role in the energy generation portfolio in the next decades despite the increasing effort on the development of renewables. In particular, coal will still be a primary energy source around the world: for example in China the coal demand is expected to reach 3.9–4.3 billion tonnes of coal by 2025 [3]. In this scenario, Carbon Capture and Storage (CCS) is one of the main candidate solutions to reduce CO_2 emissions from fossil fuel power plants. In this work post-combustion carbon capture solutions for coal-fired power plants using a hybrid membrane technology are considered and evaluated in terms of energy requirements and costs.

Solvent absorption is regarded as a mature technology for post-combustion carbon capture with amine representing the standard case for comparison. The drawback still lies in the energy penalty due to the reboiler duty in the regeneration column with energy consumption in the range 4–6 MJ/kgCO_2 captured [4,5]. Research effort have been focused on the development of new solvents that decrease the energy penalty of the integrated process [6] and on improvement of heat integration and new process configurations [6,7] that reduce energy consumptions. Membrane gas separations are already applied to air separation and natural gas sweetening

* Corresponding author.
E-mail address: M.Ferrari@ed.ac.uk (M.-C. Ferrari).

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[8]. Membrane separation processes provide a modular and flexible solution, where no solvents are needed and may represent an alternative for post combustion carbon capture from coal-fired power plants [9,10]. The low CO_2 concentration in the flue gas and the low selectivity of commercial materials are still critical and hinder the large deployment of the technology. The effect on membrane performances of other components including water vapour or impurities such as sulphur compounds, ashes and NOx is also very important. Tests on real flue gases are currently reported in order to evaluate stability and durability of the materials in the presence of contaminants [11,12].

Several papers in the literature have looked at the integration of membrane processes in power plants looking at both process and material aspects. One of the main conclusions is that with the current materials no one-stage membrane process could achieve the 90% recovery at 95% purity usually prescribed [13,14]. Two-stage processes have been analysed in depth with various configurations proposed [15–24] starting from simple cascade schemes [18] with no recycles. In addition to designs presenting only membrane stages (and a compression system), different studies show hybrid solutions where an additional separation process or a refrigeration stage is added in order to increase the efficiency of the design [14]. Highly integrated configurations which involve the recycle of part of the CO_2 to the boiler with the combustion air and therefore significant modifications of the original power plant are also considered [14,23,25–27].

Recently Roussanaly et al. [28] analysed 1600 different combinations of permeability and selectivity from the Robeson plot for the CO_2/N_2 separation in order to design the optimised simple two-stage process for each membrane and compare them with the reference amine process. Their analysis was based on a simple cascade scheme with no recycles and they were able to identify the condition for which membranes could compete with the traditional process in terms of costs. The minimum value of selectivity required for a given permeability to have an economically viable process was identified (e.g. 40 at 3500 GPU), but their analysis was limited to a binary flue gas and a simple cascade scheme; as stated in the paper, more advanced designs are expected to improve significantly the prospects for membranes.

On the materials side, research has been focused on achieving high CO_2 permeabilities and high selectivities over the other components in the flue gas [29–31]: this would guarantee an efficient separation lowering both membrane areas and energy consumptions. Recently several materials with high permeability have been reported in the literature, either polymers of intrinsic microporosity (PIMs) or Thermally Rearranged materials (TRs) [31]. Most of the process options analysed previously are based on very different transport properties, therefore process configurations specific to the novel polymers have to be devised, making sure that appropriate models are used in the simulations [32]. Additionally, since most of these novel materials are glassy polymers, their stability to ageing, contaminants and humid conditions [11–13] has become an important aspect to investigate.

In this paper, an advanced hybrid two-stage membrane carbon capture process is designed based on the transport properties of a high permeability polymer (PIM-1). Potential improvements to the process design are discussed and the effect on the process of the decline in transport properties due to ageing and the presence of impurities in the flue gas is also evaluated. The proposed process designs are linked to a rigorous economic evaluation that follows methodologies proposed by various international organisations [33–35].

2. Methodology

2.1. Power plant and solvent carbon capture process

A 550 MW coal-fired power plant with a sub-critical steam cycle (16.7 MPa/838.8 K/838.8 K) as reported by Ahn et al. [7] based on the assumptions of the 2007 DOE report, Case 9 [36] is considered as reference case (Fig. 1). The characteristics of the flue gas emitted from the power plant are reported in Table 1. The reference capture technology is an amine absorption process based on DOE Case 10 [36] using Mono Ethanol Amine (MEA) capture as retrofit. After the Flue Gas Desulfurization (FGD), the flue gas is sent to a direct contact cooler (DCC), pressurized to 131 kPa and sent to the amine absorber (30% wt MEA).

A multi-stage compression system with intercooling at 45 °C based on the DOE guidelines [36] is also included in order to have the high-purity supercritical CO_2-rich stream at 150 bar. A value of 80% [14] is assumed for the efficiency of compressor, expanders and pumps. The power required by the capture and compression sections reduces the performance of the power plant. The common assumptions for the compared process simulations are a CO_2 recovery of 90% and a final purity above 95%, as stated by the US Department of Energy guidelines [37]. All the simulations are performed using Honeywell UniSim Design® (version R400).

2.2. Membrane capture process

2.2.1. Membrane materials

In order to simulate highly permeable materials as PIM-1 (Membrane 1) a CO_2 permeance value of 3500 GPU is assumed; the value was calculated from the permeability reported for PIM-1 (7000 Barrer) [12], assuming that at this high value the support might start playing a role decreasing the permeability. The scaling factor was based on a recent report on the permeance of hollow fibres from highly permeable TR polymers that have similar high permeability to PIM-1 as thick membranes [38,39]. CO_2 selectivities over nitrogen, oxygen and water are assumed as reported in literature for flat membrane sheets [12,40]. The values of permeance and selectivity considered are reported in Table 2. In addition to the pristine PIM-1 material, two other cases

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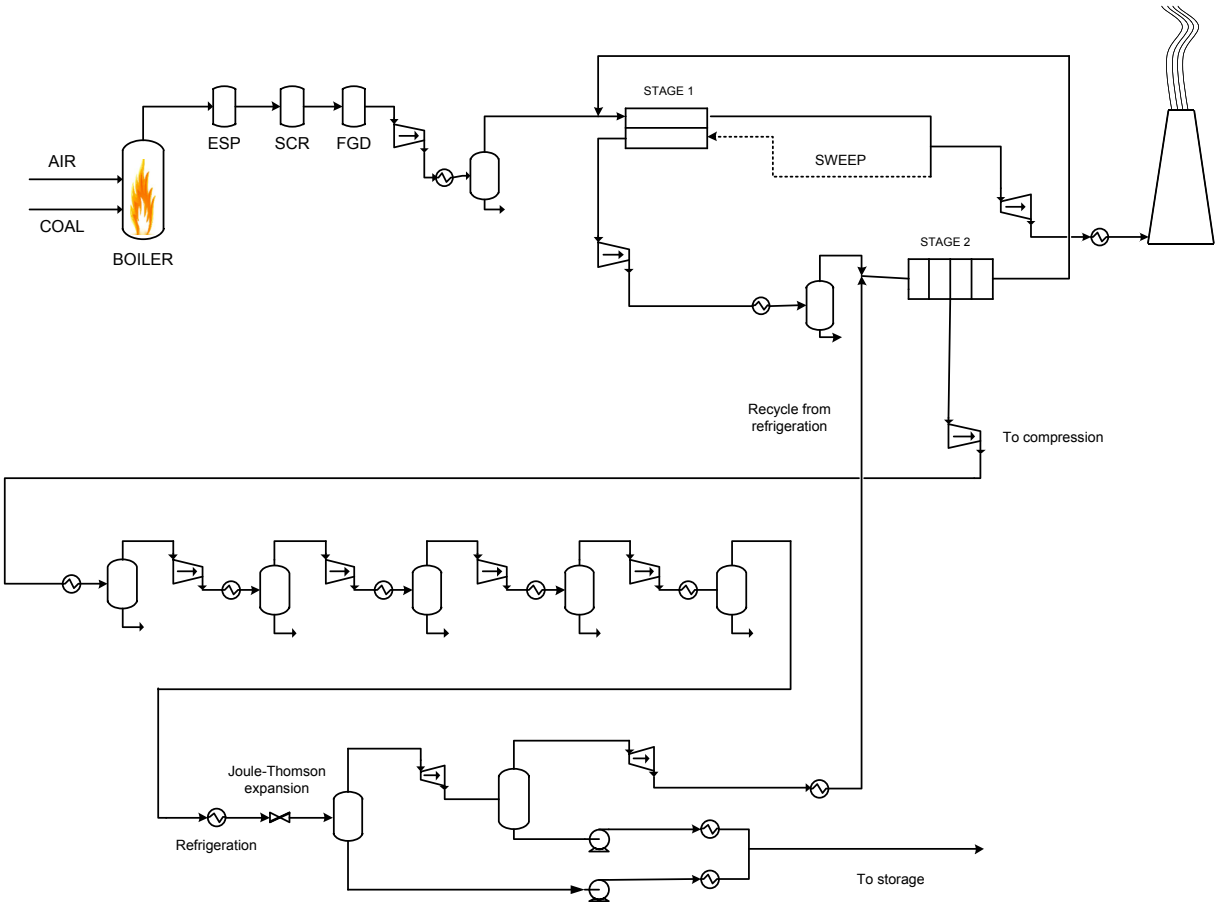

Fig. 1. Proposed retrofit configuration with optional retentate sweep (dotted line).

Table 1
DOE Case 9 flue gas conditions [37].

Molar flow rate	$2.26 \cdot 10^4$ mol/s
Molar fractions	0.13 CO_2, 0.67 N_2, 0.03 O_2, 0.17 H_2O
T and P	57 °C and 1 atm

Table 2
Membrane characteristics.

	CO_2 permeance [GPU]	Selectivity CO_2/N_2	Selectivity CO_2/O_2	Selectivity CO_2/H_2O
Membrane 1 (PIM-1) [12]	3500	19	4 [40]	1
Membrane 2 (Aged PIM-1) [41]	1300	16	4	1
Membrane 3 (PIM-1/PAF-1) [41]	3250	29	4	1

were investigated. Membrane 2 represents the case of a PIM-1 with degraded properties due to ageing. In the case of physical ageing, the permeability was decreased by 83% over 400 days and this value is used to calculate the new permeance [41]. Similar decrease has been observed in the case of exposure of the polymer to the impurities in the flue gas (i.e. NOx and SOx) [12]. Such a degradation of pristine PIM-1 is not acceptable from a process point of view and significant effort is currently devoted to make the membranes "immortal" [41]. One strategy that has proved successful is the development of mixed matrices incorporating Porous Aromatic Frameworks (PAFs) into PIM-1. Lau et al. [41] reported that the drop in permeability over time for PAF-1 loaded PIM-1 is reduced to 7% over 400 days compared to the 83% for pristine PIM-1. The addition of the porous structure not only stabilises permeability but also has a beneficial effect on selectivity over nitrogen that increases to 29. Membrane 3 represents the case of such an aged PIM-1 with Porous Aromatic Frameworks (PAFs) incorporated in order to decrease the ageing.

2.2.2. Membrane module

The membrane modules are simulated using an in-house detailed membrane module simulator implemented in UniSim Design®, with the membrane units as completely automated customised unit operations. The one-dimensional model [42] assumes a square cross-section and the hollow-fibre module geometry is described by the external dimensions (module length L, width W and height H), the fraction of volume occupied by the fibres Δ, the fibre internal r_{in} and external radius r_{out}, and consequently the thickness of the porous support θ assuming a membrane with asymmetric structure. The selective layer is assumed in the external part

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of the fibre and the feed enters the module on the shell side. Both feed and permeate sides are considered as non-dispersed plug-flow, with the option of co- and counter-current flow pattern and sweep at the permeate side. Pressure drops are included into the model formulation on both sides since their effects are expected to affect the energy consumption. The Hagen–Poiseuille relationship is assumed for pressure drops with an equivalent shell radius r_{eq} used for the shell side calculation. The equivalent radius is calculated following the relationships developed for heat exchangers [43]. Non-ideal behaviour is also taken into account using the Peng-Robinson [44] equation of state throughout the simulation. The membrane module is assumed isothermal since the process is at low pressure and the temperature difference would be negligible [45]. The numerical resolution is based on Orthogonal Collocations on Finite Elements Method (OCFEM) [46]. The set of differential equations is reduced to a system of nonlinear algebraic equations and solved with the SUNDIALS libraries [47]. The modules simulated are based on data reported in the literature (Table 3). The model was validated on the experimental data on a module presented by Woo et al. [38] with cylindrical geometry. For the full scale simulation the module dimension are retrieved from Dong et al. [32].

2.2.3. Process configuration

The hybrid two-stage configuration for the membrane capture process is shown in Fig. 1: the capture section is retrofitted to the plant without modifying it and, as for the amine case, the electrical power required for separation and compression of the CO_2 is provided by the power plant. As reported by Roussanaly et al. [28], the purity (>95%) and recovery (90%) required are not achievable with a simple two-stage process and PIM-1 membranes due to the low selectivity. To overcome this limitation and increase the purity of the CO_2, a refrigeration stage is added after the compression train. The membrane separation scheme has two stages: a first counter-current stage with the possibility of using the retentate as sweep followed by a no-sweep counter-current high-purity stage. The inlet temperature to the membrane modules is 25 °C and the pressure ratio is set at 5 for both stages as assumed in several studies [11,14]. The permeate stream from stage 2 is sent to a multi-stage compression train and the additional refrigeration stage. The CO_2 rich stream is compressed and intercooled to 45 °C in four stages from atmospheric to 30 bar and then the stream is cooled to −20 °C. A Joule–Thomson valve is used to reduce the temperature to −35 °C: two separators in series are then included to drain the liquid and the resulting CO_2-rich gas stream is sent as recycle to the inlet of stage 2 to increase the CO_2 inlet composition. Liquid CO_2 is produced after the refrigeration stage and then a pump is used in order to reach the pressure (150 bar) required for storage. An efficiency of 50% for the refrigeration cycle [48] is assumed in order to estimate the overall energy consumption.

A summary of all the simulations performed is reported in Table 4.

2.3. Economic analysis

Economic analysis of the process configuration simulated is essential to evaluate the feasibility of the proposed membrane technology in comparison with other solutions: different international organisations report detailed guidelines for carbon capture methodologies and cost targets [33,34,49]. To estimate the impact of the capture system on the plant performances, the cost of electricity is used and in particular Levelised Cost Of Electricity (LCOE) based on the methodologies adopted by UK Department of Energy and Climate Change (DECC) [35,49] and European Zero Emission Platform (ZEP) [33]. The LCOE is ratio of the net present value of total capital and operating costs of a plant to the net present value of the net electricity generated by that plant over its operating life [49]. The formulation from DECC is given by (1) with: I_t the total overnight capital cost, M_t the sum of the fixed/variable annual operating costs, E_t the annual output at 100% capacity and F_t is the fuel cost and r is the discount rate.

$$LCOE = \frac{\sum_{t=1}^{n} \frac{I_t + M_t + F_t}{(1+r)^t}}{\sum_{t=1}^{n} \frac{E_t}{(1+r)^t}} \quad (1)$$

Capital and O&M costs included in I_t and M_t are calculated according to the formulation reported by Zhao et al. [18] for

Table 3
Module characteristics.

	Module for model validation (Woo et al. [38])	Module for process simulation (Dong et al. [32])
Length of the module [m]	0.15	0.915
Width of the module [m]	0.067	0.135
Height of the module [m]	0.067	0.135
Fibre fraction [-]	0.00039	0.392
External radius of the fibre [m]	3.60E-04	3.60E-04
Ratio between internal and external radius [-]	0.771	0.771
Total number of fibres [-]	4	17500
Area [m^2]	1.48E-03	36.17

Table 4
Overview of configurations presented in this paper.

Simulation	Short description
Base Case	Base power plant [36]
Amines	Solvent capture, DOE Case 10 [36]
Case A	Membrane 1 (no retentate sweep)
Case B	Membrane 2 with process design of Case A
Case C	Membrane 3 with process design of Case A
Case D	Membrane 2
Case E	Membrane 3
Case F	Membrane 1, permeate pressure of stage 1 fixed to 500 mbar and no retentate sweep.
Sweep 1–5	Configuration with retentate of stage 1 as sweep: feed pressure fixed to 1.1 bar and membrane 1 used

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their carbon cost estimation. They are reported in (2) and (3), where:

- Suffixes O&M refer to Operation and Maintenance contributions;
- I_c, I_{vp}, I_{ex}, I_{ref}, I_{he}, I_m and I_{mf} are respectively the costs of compressors, vacuum pumps, expanders, refrigeration, heat exchangers, membranes and membrane frames;
- a and a_m represent annualised factors for equipment and membrane contributions;

$$C_{cap} = a\left(I_c + I_{vp} + I_{ex} + I_{he}\right) + a_m I_m \quad (2)$$

$$C_{O\&M} = 0.036\left(I_c + I_{vp} + I_{ex} + I_{he}\right) + 0.01\left(I_m + I_{mf}\right) \quad (3)$$

The complete list of parameters used in the LCOE calculation is reported in Table 5.

In the case of power plants with carbon capture, the contribution of capital, operating and maintenance (O&M) and storage are added to the base case calculation. Therefore, terms, in I_t and M_t in (1) include contribution of the capture process. The parameters from the capture cost estimation from Zhao et al. [20] are adopted for the membrane contribution, considering the O&M the same escalation factor as fuel (1.5%). It is assumed that the membranes are changed every 5 years while the skids and the equipment twice in the life of the plant (an average life of 25 years for the membrane skids is assumed [20]).

The cost parameters for the base case plant are assumed as nth of a kind (NOAK) coal-fired power plant [35]. The choice of using NOAK parameters implies that the technology in the field is already mature and a number of plants are built, which is a reasonable assumption in the case of a coal-fired power plant with FGD. Capital costs include the contributions of pre-licensing, regulatory, Engineering and Procurement Costs (EPC) and infrastructure costs. Operation and maintenance include fixed and variable fees accounting for an annual escalation factor of 2% and an average fuel cost estimation is assumed [33]. The energy penalty for the amine reference case is 135 MW, 24.4% of the net power output. The cost parameters for the LCOE calculation based on amine capture are considered as 1st of a kind amine technology [49]. Transport and storage costs are evaluated according to UK DECC guidelines [49] and are assumed equal for both amines and membrane technology. In the case of unknown parameters for the membrane case (for example EPC and licensing costs), they are taken from the amine analysis as 1st of a kind (FOAK). In this case the assumption of FOAK parameters is justified since LCOE is predicted for a new-built plant with carbon capture. The LCOE methodology and the possible contribution of carbon cost have been investigated in the European environment. Emission Trading Scheme (ETS) [50] has the potential of giving an important contribution to the deployment of the technology reducing the costs. Each plant has an assigned amount of emissions according to the current European regulation [50]: the mechanism is based on a cap-and-trade system, which considers additional emissions as a debit. The integration of CCS in the ETS system is still under development, but the captured CO_2 can be considered as benefit for the plant owner in terms of European Unit Allowances (EUAs) and it has been considered in the LCOE calculation. Despite the difficulties in identifying a trend in carbon price for the next decades, an emission cost of 6 €/tCO_2 for 2016 with an escalation of 2 €/year was assumed. This can be considered a conservative assumption in comparison to available reports [33,49] and predictions [51]. In order to integrate the possible role of the emission market, a carbon tax scenario consistent with the UK DECC analysis was also assumed in this paper.

Table 5
Parameters used for the LCOE calculation.

General	Base	CCS
Plant life [35] [years]	40	
Discount rate [35] [-]	0.1	
Fuel		
Fuel cost [33] [€/GJ (LHV)]	2.4	
Fuel escalation cost (year) [33][-]	0.015	
Capital costs		
Pre-licensing costs [35] [£/kW]	25	25
Regulatory + licensing [35] [£/kW]	0.2	1.5
EPC cost [33] [£/kW]	1555	2450
Infrastructure cost [35] [£/kW]	22.5	22.5
O&M		
O&M fixed fee [35] [£/MW/y]	35000	35000
O&M variable fee [35] [£/MWh]	1	2.5
O&M escalation cost (year) [35] [-]	0.02	0.02
Insurance [35] [£/MW/y]	2500	4400
Connection and UoS charges [35] [£/MW/y]	5050	5050
Storage		
CO_2 transport and storage [35] [£/MWh]	0	7.8

3. Results and discussion

3.1. Model validation

In order to validate the implemented model, the conditions explored by Woo et al. [38] are replicated. This was chosen as a reference because it is focused on a highly permeable polymer similar to what is considered in this study and uses similar contributions in the model (i.e: pressure drop on the

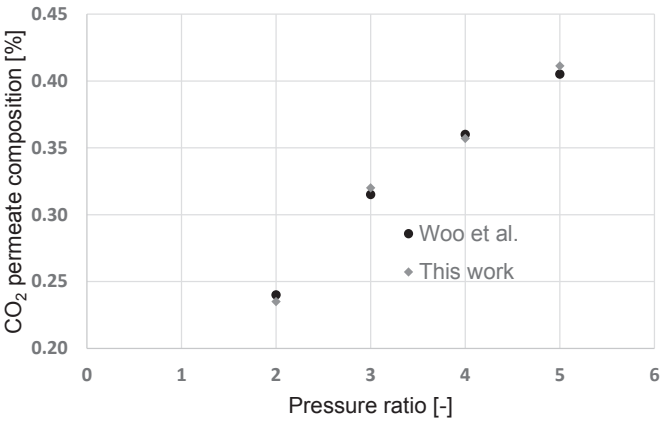

Fig. 2. Comparison of results with literature data [32].

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Table 6
Summary of cases simulated with no sweep.

Simulation	Material	Area 1 [10^6 m^2]	Area 2 [10^6 m^2]	Total area [10^6 m^2]	Energy consumption [MW]	Recovery [%]	Purity [%]
Case A	Membrane 1	1.37	0.34	1.72	200	90%	98.7%
Case B	Membrane 2 with Design 1	1.37	0.34	1.72	102	55%	98.8%
Case C	Membrane 3 with Design 1	1.37	0.34	1.72	161	87%	98.7%
Case D	Membrane 2	3.26	0.89	4.14	206	90%	98.7%
Case E	Membrane 3 with new design	1.54	0.39	1.93	174	90%	98.7%

Table 7
Energy consumption comparison of Case A vs. solvent capture.

Configuration	Net power output [MW]	Energy consumption for capture unit [MW]	Energy consumption for compression [MW]	Energy consumption for refrigeration [MW]
Base Case	550	–	–	–
Case A	350	110	74	16
Amines	418	100	32	–

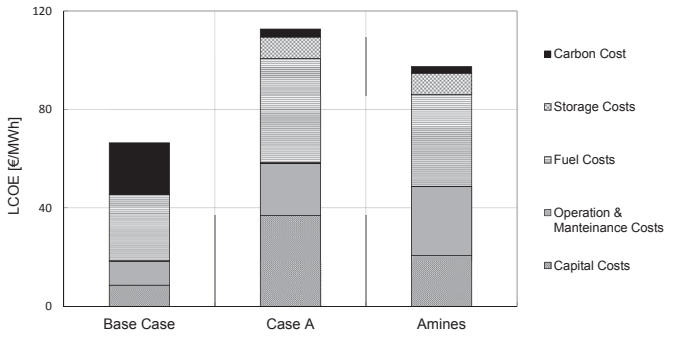

Fig. 3. Levelised cost of electricity for two-stage membrane process with membrane 1 (Case A) compared to the reference power plant and amine capture cases.

permeate side as discussed by Dong et al. [32]). Fig. 2 shows the comparison between the experiments reported in the literature and our simulation and the two are in perfect agreement further demonstrating that the square cross-section assumed has no influence on the results.

3.2. Base membrane case

As described in Section 2.2.1, the advanced hybrid configuration chosen includes two membrane stages and a refrigeration stage at the end of the capture process in order to achieve high purity. The base membrane configuration (Case A) includes recycles but no sweep in the two membrane modules. The results in terms of area and energy requirement are reported in Table 6 while the breakdown of the energy consumption is summarised in Table 7. The results show that it is possible to design an advanced two-stage process with high permeability materials with modest selectivity as hinted by Roussanaly et al. [28]. Fig. 3 shows the comparison in terms of economics with the reference amine case. As expected, the process is not competitive in terms of LCOE with the state of the art amine process and this is due to the low selectivity; further advanced configuration need to be explored for this type of materials.

Table 8
Energy consumption and membrane area comparison with same pressure ratio (2.2) with and without retentate sweep.

Simulation	Material	Area 1 [10^6 m^2]	Area 2 [10^6 m^2]	Total area [10^6 m^2]	Energy consumption [MW]	Recovery [%]	Purity [%]
Case F	Membrane 1	4.01	9.04	4.92	233	90%	98.80%
Sweep 4	Membrane 1	2.93	5.42	3.47	205	90%	98.80%

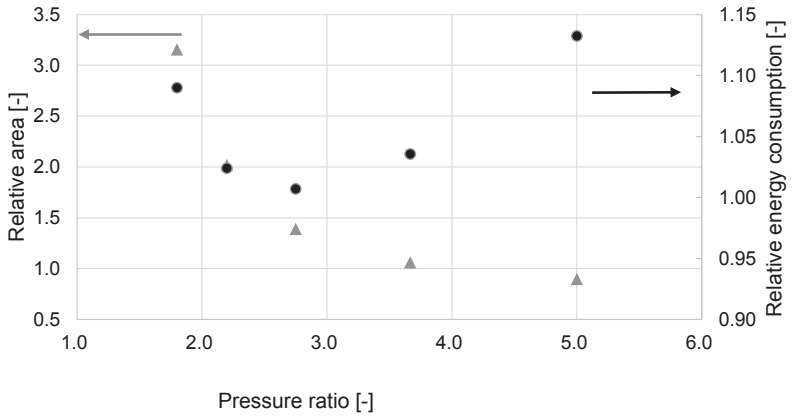

Fig. 4. Effect of sweep pressure (reported as pressure ratio with fixed feed pressure at 1.1 bar) on total membrane area and energy requirements relative to Case A. The fraction of retentate used as sweep is reported in Table 9 for each point.

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Table 9
Overview of sweep configurations with respective pressure ratio and sweep-to-retentate ratio.

Configuration name	Pressure ratio	Retentate sweep fraction
Sweep 1	5.0	18%
Sweep 2	4.0	18%
Sweep 3	3.0	20%
Sweep 4	2.2	27%
Sweep 5	1.8	39%

3.3. Effect of sweep pressure

One option in order to reduce the area of the membranes and therefore the really high capital cost of the process is to add a sweep stream. Our choice was to use part of the retentate as sweep stream following the results of Merkel et al. [14] that demonstrated the improvement in the area compared to only vacuum on the permeate side. This would clearly increase the complexity of the layout of the plant and the option has to be economically advantageous in order to be attractive. To confirm the effect of sweep at low pressure ratios, simulations have been carried out with and without sweep by fixing permeate pressure of stage 1–500 mbar (pressure ratio stage 1 fixed to 2.2). The results are reported in Table 8, where both a dramatic reduction of membrane area (42%) and energy consumption (14%) can be observed if retentate sweep is used.

Fig. 4 reports the results for different pressure ratios in the first stage, keeping fixed the feed pressure; as expected the area required for the separation decreases dramatically increasing the level of vacuum while the energy required has a minimum around a pressure ratio of 2.8 in the first stage. An overview of the configurations is also presented in Table 9 where the fraction of retentate used as sweep is reported.

In Fig. 5 the LCOE and its breakdown is compared to Case A for membranes showing that the sweep option becomes competitive at pressure ratios between 2.5 and 4. This is due to the dramatic reduction of membrane area compared to lower pressure ratios that compensate for the higher energy requirements. If the sweep pressure if further decreased (i.e: increasing the pressure ratio) the reduction in the area and membrane capital cost is not enough to compensate the increase in vacuum requirement of the plant.

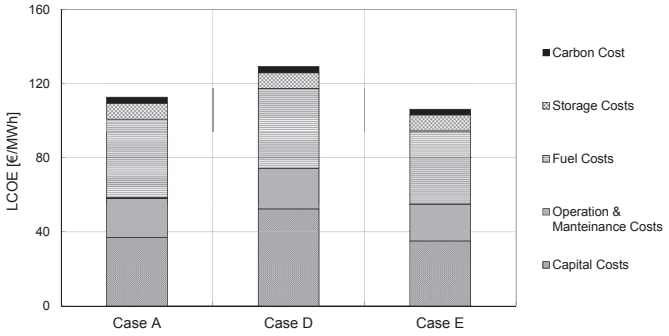

Fig. 7. Comparison of optimised configuration costs for Cases A, D and E.

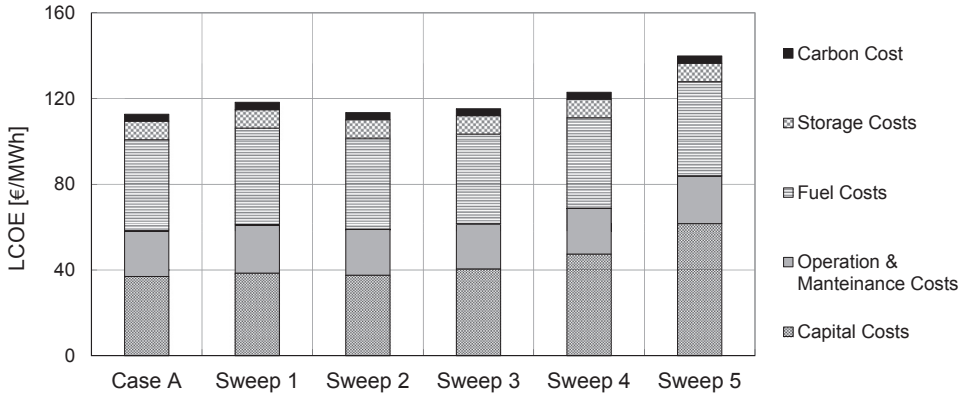

Fig. 5. LCOE and breakdown of contribution for base membrane case and alternative sweep configurations (listed in Table 9).

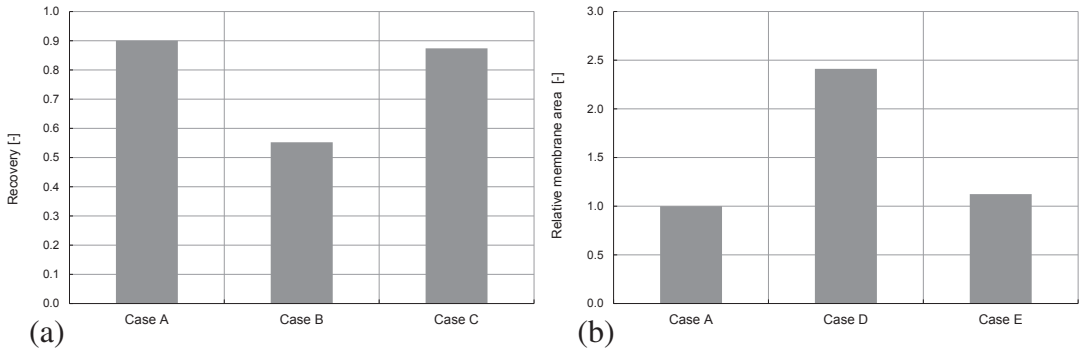

Fig. 6. Effect of material ageing and degradation (a) on the process recovery for the same design of Case A and (b) on the membrane area at fixed 90% recovery.

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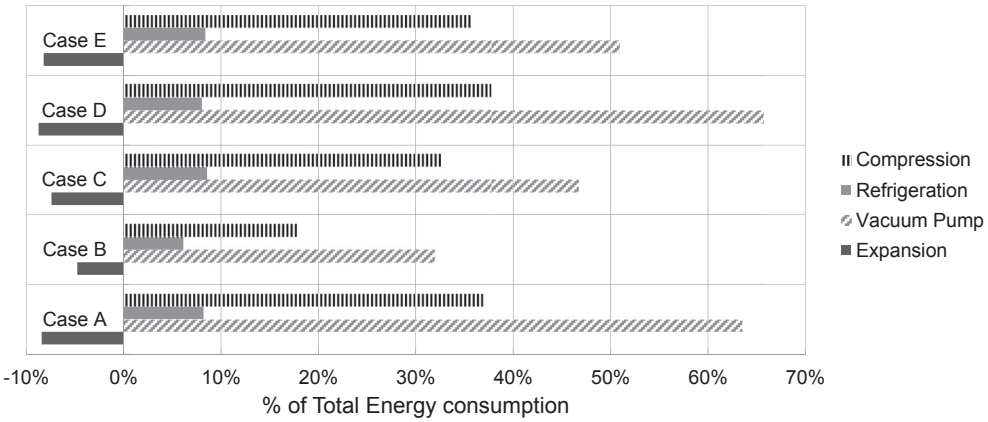

Fig. 8. Breakdown of power consumption for the different cases.

3.4. Effect of impurities and ageing effect

One of the main problems related to super glassy polymer is their tendency to age over time and be affected by exposure to water and impurities [12]. In order to predict the performance of a fixed design over the life of the membrane, the base design for PIM1 was evaluated for a membrane with the properties after ageing or after exposure to impurities. The properties considered for the degraded materials are reported in Table 3. As it can be seen in Fig. 6 the recovery drops dramatically once the degraded PIM-1 is considered while the purity is not affected due to the presence of the cryogenic stage. The operating cost of the original plant with the degraded membrane will vary due to the reduced flow rates and therefore affect the LCOE that would also increase significantly due to the carbon cost. This simulation also confirms that in case of a capture regime below 90% recovery, membranes can have a significant advantage compared to other technologies even for modest selectivity. A capture process can also be designed for a 90% recovery with the properties of the degraded PIM-1 (Case D) with an expected increase in membrane area required and energy consumption that lead to a dramatic increase in the LCOE (Table 3).

Using the properties of a PIM-1 stabilised with PAFs (Membrane 3) to evaluate our base membrane design will instead results in a drop in recovery to just 88% not far off the 90% target (Case B). A process with 90% recovery can be designed for the PIM-1-PAF materials. This would lead in a 22% overdesign of the membrane modules compared to the base case as reported in Table 3 (Case E). The LCOE (Fig. 7) will be lower than the base case due to the higher selectivity and it is expected that process improvement similar to the sweep cases considered in the previous section will lead to even more competitive processes.

Finally the breakdown of the power consumption for the different simulation is shown in Fig. 8 and confirms the advantage of the capture configuration including a refrigeration stage. In all cases the contribution of the refrigeration step to the energy requirement is relatively small while in terms of purity it allows to move from around 80% out of the second membrane stage to over 98%.

4. Conclusions

The development of super glassy polymers with really high permeability but limited selectivity requires the design of specific advanced process configurations that take advantage of their unique properties and remain economically viable. These materials are attractive for post combustion capture application due to the large volume of flue gas to be treated. Simulations of a hybrid two-stage configuration show that membranes with really high permeability like PIMs or TR polymers can deliver the recovery and purity required for storage but are not yet competitive in terms of economics. More advanced process configurations should be investigated in order to lower the energy and the area required for the separation bringing LCOE below 100 €/MWh, in line with more mature technologies. Finally the degradation of high performance materials due to ageing and interaction with impurities has a large impact on the process recovery and economics and should be taken into account in evaluating any design. Efforts in testing materials close to real conditions are necessary in order to produce a reliable estimation of future plant performances.

Conflict of interest

The authors declare no conflict of interest.

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Appendix.

A more detailed representation of the proposed membrane design is reported in Fig. A.

In Table A temperature, pressure, flow and composition in each stream can be found. This is reported as representative case for Case A.

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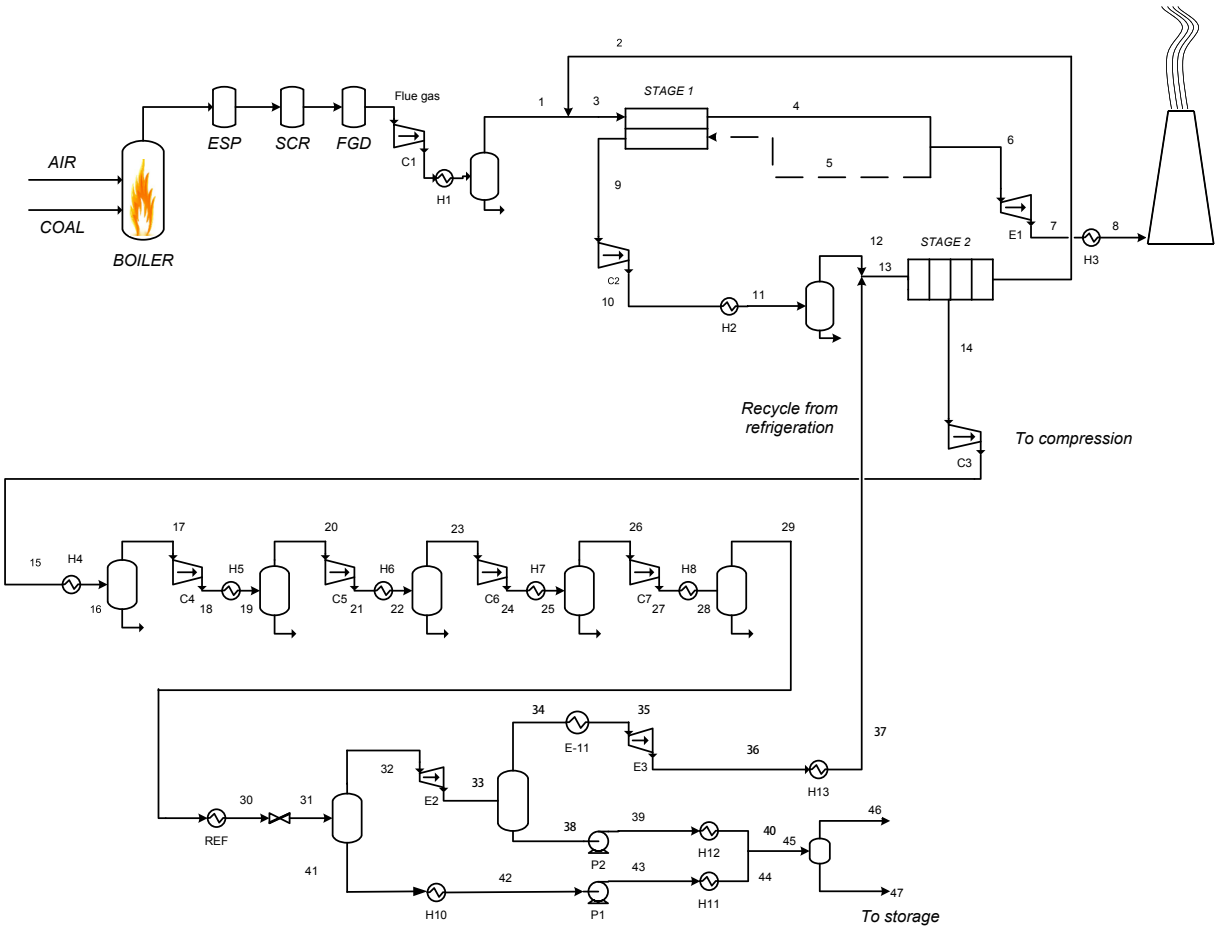

Fig. A. Detailed representation of the design reported in Fig. 2.

Table A
Breakdown of pressure for each stream in Case A.

Stream	Flue gas	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Flow [mol/s]	2.02E + 04	1.87E + 04	1.24E + 04	3.11E + 04	1.56E + 04	1.56E + 04	1.56E + 04	1.56E + 04	–	–	–	1.55E + 04	1.55E + 04	1.55E + 04	1.53E + 04	1.81E + 04	5.72E + 03
T [K]	330	298	298	298	298	298	298	323	–	–	–	298	475	298	298	298.1	298.1
P [MPa]	0.100	0.110	0.110	0.110	0.109	0.101	0.101	0.101	–	–	–	0.022	0.110	0.110	0.110	0.110	0.020
O_2	2.1%	2.2%	8.4%	4.7%	2.6%	2.6%	2.6%	2.6%	–	–	–	6.90%	6.90%	6.90%	6.9%	7.8%	6.30%
N_2	73.6%	79.5%	59.2%	71.5%	95.4%	95.4%	95.4%	95.4%	–	–	–	47.40%	47.40%	47.40%	48.1%	43.6%	9.90%
H_2O	9.5%	2.3%	1.4%	1.9%	0.1%	0.1%	0.1%	0.1%	–	–	–	3.70%	3.70%	3.70%	2.5%	2.1%	3.60%
CO_2	14.8%	2.3%	31.0%	21.9%	1.9%	1.9%	1.9%	1.9%	–	–	–	42.00%	42.00%	42.00%	42.5%	46.5%	80.20%
Total	100.0%	86.3%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Stream	**17**	**18**	**19**	**20**	**21**	**22**	**23**	**24**	**25**	**26**	**27**	**28**	**29**	**30**	**31**	**32**	**33**
Flow [mol/s]	5.72E + 03	5.72E + 03	5.72E + 03	5.72E + 03	5.72E + 03	5.67E + 03	5.67E + 03	5.67E + 03	5.61E + 03	5.61E + 03	3.38E + 03	5.56E + 03	5.56E + 03	5.56E + 03	5.56E + 03	5.56E + 03	5.56E + 03
T [K]	298.1	298.1	298.1	298.1	318.1	318.1	399.15	318.1	318.1	400.5	318.1	318.1	399.8	323.1	323.1	253.1	243.6
P [Pa]	0.100	0.100	0.100	0.240	0.24	0.24	0.56	0.56	0.56	1.32	1.32	1.32	3.12	3.12	3.12	3.12	2.00
O_2	6.30%	6.30%	6.30%	6.30%	6.30%	6.40%	6.40%	6.40%	6.40%	6.40%	6.40%	6.50%	6.50%	6.50%	6.50%	6.50%	6.50%
N_2	9.90%	9.90%	9.90%	9.90%	9.90%	10.00%	10.00%	10.00%	10.20%	10.20%	10.20%	10.20%	10.20%	10.20%	10.20%	10.20%	10.20%
H_2O	3.60%	3.60%	3.60%	3.60%	3.60%	2.70%	2.70%	2.70%	1.60%	1.60%	1.60%	0.76%	0.76%	0.76%	0.76%	0.76%	0.76%
CO_2	80.20%	80.20%	80.20%	80.20%	80.20%	80.90%	80.90%	80.90%	81.80%	81.80%	81.80%	82.50%	82.50%	82.50%	82.50%	82.50%	82.50%
Total	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Stream	**34**	**35**	**36**	**37**	**38**	**39**	**40**	**41**	**42**	**43**	**44**	**45**	**46**	**47**			
Flow [mol/s]	2.94E + 03	2.94E + 03	2.80E + 03	2.80E + 03	2.80E + 03	1.37E + 02	1.37E + 02	3.26E + 01	2.62E + 03	2.67E + 03	2.67E + 03	2.76E + 03	1.50E + 02	2.71E + 03			
T [K]	243.6	222.94	222.94	195.9	298.1	222.94	229.5	298.1	243.6	249	298.1	298.1	298.1	298.1			
P [MPa]	2.00	1,00	1,00	0.12	0.12	1,00	15,00	15,00	2,00	15,00	15,00	15,00	15,00	15,00			
O_2	11.70%	11.70%	12.30%	12.30%	12.30%	0.32%	0.32%	0.32%	0.60%	0.60%	0.60%	0.60%	0.00%	0.58%			
N_2	18.70%	18.70%	19.60%	19.60%	19.60%	0.35%	0.35%	0.35%	0.70%	0.70%	0.70%	0.70%	0.00%	0.70%			
H_2O	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	1.60%	1.60%	1.60%	1.50%	100.00%	0.00%			
CO_2	69.60%	69.60%	68.10%	68.10%	68.10%	99.30%	99.30%	99.30%	97.10%	97.10%	97.10%	97.20%	0.00%	98.70%			
Total	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%			

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